

U.S. Serial Number: 09/553,107  
Reply to Office Action of: 12/27/2007  
Family Number/Docket Number: P1998J107D/GJH-0018

Page 6 of 13

RECEIVED  
CENTRAL FAX CENTER

APR 23 2008

### REMARKS

Applicants thank the Examiner for the interview of March 10, 2008. The contents of the interview are summarized by the remarks below.

Claim 1 is amended to require that the distillate feedstock contains polynuclear aromatics, and to require that the liquid product stream of step (d) has a weight ratio of aromatics to polynuclear aromatics of at least about 11. Support for the amendments to claim 1 can be found, for example, at the top of page 14 of the specification as filed, and in Figure 2.

Claim 1 is also amended to replace the transitional phrase "consisting essentially of" with "comprising". The transitional phrase "comprising" was present in claim 1 as originally filed. The choice of transitional phrase does not impact the distinctions provided in the remarks below with regard to demonstrating the patentability of the claimed invention.

#### **I. Rejections under 35 U.S.C. §103**

The rejection of claims 1 – 7, 9 – 12, 16, and 18 under 35 USC 103(a) over Harrison et al, US 5,292,428 is respectfully traversed. Applicants have unexpectedly found that the amount of polynuclear aromatics in a distillate fuel product can be substantially reduced by limiting the hydrogen portion of the treat gas in the second hydrodesulfurization stage to less than or equal to three times the chemical hydrogen consumption.

U.S. Serial Number: 09/553,107  
Reply to Office Action of: 12/27/2007  
Family Number/Docket Number: P1998J107D/GJH-0018

Page 7 of 13

Distinctions between total aromatics vs. polynuclear aromatics

Polynuclear aromatic compounds, also referred to as PNAs, are an undesirable species in a distillate fuel product such as diesel. As noted on page 14 of the specification as filed, PNAs are aromatic compounds that have two or more aromatic rings. PNAs contribute significantly to the emissions produced by diesel engines, and therefore it is desirable to reduce the amount of PNAs present in a diesel fuel.

Additionally, as noted on page 2, regulations have been developed in more than one jurisdiction that provide separate limits for the amount of total aromatics and the amount of polynuclear aromatics in a diesel fuel. Thus, it is desirable to be able to separately control the amount of polynuclear aromatics relative to the amount of total aromatics.

Conventional methods of hydrotreatment are not selective for removing PNAs relative to other aromatic compounds. Attempting to simply hydrogenate all aromatics within a distillate fuel product would lead to a large level of hydrogen consumption. Such a level of hydrogen consumption would substantially increase the cost of production for a distillate fuel product.

The claimed invention – Solving the problem of separately controlling the level of polynuclear aromatics relative to total aromatics

The claimed invention provides a method for balancing the competing interests identified above. By maintaining a hydrogen concentration in a second hydrotreatment stage as described in the claims, the level of PNAs within a distillate fuel product can be reduced without excessive hydrogen consumption. A convenient metric for understanding whether PNAs have been reduced without excessive hydrogen

U.S. Serial Number: 09/553,107  
Reply to Office Action of: 12/27/2007  
Family Number/Docket Number: P1998J107D/GJH-0018

Page 8 of 13

consumption is to determine the ratio of total aromatics to PNAs within the resulting distillate fuel product. If PNAs remain high, this ratio will have a low number. On the other hand, if aromatics are indiscriminately hydrogenated, this ratio will also have a low number. A high ratio value is only achieved by reducing the number of PNAs while minimizing the hydrogenation of other aromatics.

The benefits of the invention can be understood based on the data contained in Tables 1 and 2. Table 1 presents product properties for feeds processed according to the invention. For each product in Table 1, a distillate fuel product is produced with a total sulfur level below 100 wppm and a ratio of aromatics to PNAs of greater than 13. By contrast, Table 2 provides comparative examples of distillate fuel products produced by other methods. The highest ratio of aromatics to PNAs in Table 2 is 10.45, for the process used in Comparative Example C.

The distinction between the claimed invention and the Comparative Examples is readily seen in Table 2. Table 2 plots the ratio of aromatics to PNAs for each of the inventive examples from Table 1 and each of the Comparative Examples from Table 2.

The benefits of the claimed invention are achieved by controlling the rate of hydrogen introduction in step (d)

Part of Applicants' discovery is that selective hydrogenation of polynuclear aromatics can be achieved by limiting the amount of hydrogen present in a second (or later) hydrodesulfurization stage. As required in step (d) of claim 1, the rate of introduction of the hydrogen portion of the treat gas in this second stage is less than or equal to 3 times the chemical hydrogen consumption in this second reaction stage.

U.S. Serial Number: 09/553,107  
Reply to Office Action of: 12/27/2007  
Family Number/Docket Number: P1998J107D/GJH-0018

Page 9 of 13

The Harrison reference does not describe control of polynuclear aromatic levels

The Harrison reference provides no discussion of a distinction between PNAs and other aromatic compounds. Instead, Harrison appears to be directed to solving the problem of providing an improved hydrodesulfurization process that reduces the total aromatics levels within a feedstock. (See, e.g., Harrison, Col. 7, lines 25 – 31.) Because Harrison is focused on reduction of the amount of total aromatics, Harrison fails to even identify the problem of reducing the amount of PNAs preferentially relative to other aromatic compounds.

The method for introducing hydrogen into the reaction system in Harrison differs qualitatively from the method of hydrogen introduction in the claimed invention

In part because of Harrison's different focus, Harrison does not have any discussion or suggestion of limiting hydrogen introduction in the second hydrotreatment stage as required by the claimed invention. Instead, due to the different focus of the invention in Harrison, Harrison requires that all makeup hydrogen is introduced into later reaction stages. In particular, new hydrogen from outside the system is not introduced into the first hydrodesulfurization stage, regardless of how many additional stages are present. According to Harrison, this distribution of hydrogen allows for the best hydrodesulfurization and aromatics saturation of a feedstock. (See Col. 10, line 33 – Col. 11, line 61 for a discussion of how to introduce hydrogen into the apparatus of Harrison and the resulting benefits for hydrodesulfurization and total aromatics saturation.)

Because Harrison does not introduce makeup hydrogen into the first hydrodesulfurization stage, all makeup hydrogen for the reaction must be introduced into a later stage. Since a large portion of the aromatic saturation in Harrison takes place in the first reaction stage, enough makeup hydrogen must be introduced into the

U.S. Serial Number: 09/553,107  
Reply to Office Action of: 12/27/2007  
Family Number/Docket Number: P1998J107D/GJH-0018

Page 10 of 13

second (or later) stages to account for the hydrogen consumption in the first stage. Thus, Harrison explicitly requires that enough excess hydrogen to satisfy hydrogen needs in the first hydrodesulfurization stage must be added to later hydrodesulfurization stages. This is in sharp contrast to the claimed invention, where hydrogen introduction into the last hydrodesulfurization stage is carefully limited based on the hydrogen consumption for that stage.

The Examples provided in Harrison do not satisfy the hydrogen introduction requirements of step (d) of claim 1

The claimed invention requires the rate of introduction of the hydrogen portion of the treat gas in step (d) to be less than or equal to 3 times the chemical hydrogen consumption in that reaction stage. The calculations below will demonstrate that in the Examples of Harrison, the amount of hydrogen introduced into the second hydrodesulfurization stage is greater than 3 times the hydrogen consumption of the entire system in Harrison. Since all of the hydrogen in Harrison is introduced into the second stage, if the amount of hydrogen in the Harrison treat gas is greater than 3 times the hydrogen consumption of the entire system, then it must follow that the amount of hydrogen in the treat gas is also greater than 3 times the hydrogen consumption in the second stage.

To demonstrate the hydrogen consumption in Harrison, Example 5 will be used as it provides the lowest ratio of treat gas provided to hydrogen consumption. In the following calculations, hydrogen consumption will be calculated using a model that matches the physical characteristics of the feed as specified in Harrison, while allowing for as much hydrogen consumption as possible.

Harrison states that the average molecular weight of the molecules in its feedstock is 365. In the following discussion all aromatics in Harrison are assumed to

U.S. Serial Number: 09/553,107  
Reply to Office Action of: 12/27/2007  
Family Number/Docket Number: P1998J107D/GJH-0018

Page 11 of 13

be 2 ring aromatics that have the average molecular weight of the feed. For aromatics that are converted into non-aromatic molecules, it will be assumed that the molecule is fully saturated. For aromatics that still remain aromatics in the product, it will be assumed that some hydrogenation still occurs, so that one of the two aromatic rings becomes saturated. It is believed that this provides the maximum hydrogen consumption, while still corresponding to the average molecular weight specified in Harrison.

Based on the above assumptions, the relative hydrogen consumption in Harrison can be calculated. In example 5, the feed provided contains 27.7 vol% aromatics while the product is 16.6% aromatics. The average feed molecular weight is specified as 365, while the density of the feed is  $944 \text{ kg/m}^3$ , which is equivalent to 944 g/liter. Based on these numbers, roughly 0.287 moles of aromatics are saturated per liter of feed.  $(944 \times 11.1/100 \times 1/365)$  As noted above, for the purpose of calculating hydrogen consumption, we will assume that all of the converted aromatics are two-ring aromatics that are converted to fully saturated two-ring naphthenes. This would require 5 moles of  $\text{H}_2$  per mole of aromatic converted, or 1.435 moles  $\text{H}_2$ . Additionally, the remaining 16.6 vol% aromatics in the product are also assumed to be saturated from a two-ring aromatic to a one-ring aromatic. This requires an additional 2 moles of  $\text{H}_2$  per mole of aromatic molecules, or 0.858 moles of  $\text{H}_2$ . Thus, the total  $\text{H}_2$  consumption in this model calculation is 2.293 moles of  $\text{H}_2$  per liter of feed. Note that any amount of hydrogen consumed by desulfurization is small relative to the hydrogen consumed by aromatic saturation.

As shown above, the total  $\text{H}_2$  consumption is 2.293 moles per liter of feed. Based on the feed flow rate provided in Example 5, this corresponds to 322 SCF/b. By contrast, the flow rate for hydrogen treat gas into the Harrison reactor is 1370 SCF/b. Thus, the ratio of hydrogen provided to hydrogen consumed in Harrison is greater than about 4.25, for the entire reactor in Harrison. Again, if the ratio of hydrogen provided

U.S. Serial Number: 09/553,107  
Reply to Office Action of: 12/27/2007  
Family Number/Docket Number: P1998J107D/GJH-0018

Page 12 of 13

to hydrogen consumed in the entire reactor is greater than about 4.25, then the ratio of hydrogen provided to hydrogen consumed in only the second reaction stage will be still higher. As a result, Harrison does not describe or suggest the claimed requirement of providing less than or equal to 3 times the hydrogen consumption of the second reaction stage.

As a final note, with regard to basis provided for the rejection in the prior Office Action: The Examiner has previously noted Col. 10, lines 3 – 11 of Harrison, where Harrison states that it is preferred to use at least 1.05 times the stoichiometric amount of hydrogen during the hydrogenation process described in Harrison, and that some additional allowance should be made for hydrogen dissolved in a recovered treated feedstock. This is merely a statement that enough hydrogen must be present allow the hydrogenation reaction to stoichiometrically occur, and provides no insight into the benefits of controlling the amount of hydrogen available during hydrotreatment.

As demonstrated above, Harrison does not describe or suggest the requirement of the claimed invention of limiting the hydrogen portion of the treat gas in the second stage to 3 times or less the chemical hydrogen consumption. Because Harrison fails to describe or suggest this claimed requirement, as well as failing to describe or suggest the unexpected benefit produced by practicing the claimed invention, claims 1 – 7, 9 – 12, 16, and 18 are patentable in view of Harrison. Reconsideration and withdrawal of the rejection are respectfully requested.

## **II. Conclusion**

Having demonstrated that all rejections of claims have been overcome, this application is in condition for allowance. Accordingly, applicants request early and favorable reconsideration in the form of a Notice of Allowance.

APR 23 2008

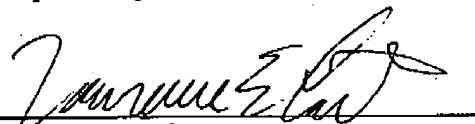
U.S. Serial Number: 09/553,107  
Reply to Office Action of: 12/27/2007  
Family Number/Docket Number: P1998J107D/GJH-0018

Page 13 of 13

If there are any questions regarding this amendment or the application in general, a telephone call to the undersigned would be appreciated, since this should expedite the prosecution of the application for all concerned.

If necessary to effect a timely response, this paper should be considered as a petition for an Extension of Time sufficient to effect a timely response. Please charge any deficiency in fees or credit any overpayments to Deposit Account No. 05-1330.

Respectfully submitted,



Lawrence E. Carter  
Attorney for Applicant(s)  
Registration No. 51,532  
Telephone Number: (908) 730-3632  
Facsimile Number: (908) 730-3649

☒ Pursuant to 37 CFR 1.34

ExxonMobil Research and Engineering Company  
P. O. Box 900  
Annandale, New Jersey 08801-0900

4/22/08